

**DU COMBUSTIBLE NUCLÉAIRE AUX DÉCHETS :
RECHERCHES ACTUELLES**
FROM NUCLEAR FUELS TO WASTE: CURRENT RESEARCH

Long-term performance of spent fuel in geological repositories

Wernt Brewitz*, Ulrich Noseck

Gesellschaft für Anlagen- und Reaktorsicherheit (GRS) mbH, Theodor-Heuss-Strasse 4,
38122 Braunschweig, Germany

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Note presented by Édouard Brézin.

Abstract

The amount of spent fuel generated in Germany up to 2040 has been estimated at 8950 t_{hm}. By this time 5500 t_{hm} of vitrified high level waste will have accumulated. Both types of waste will be finally disposed of in one underground repository. Potential host rocks are rock salt, granite and mudrock. Up to now several performance assessment studies have been carried out in order to identify the main parameters governing the long-term safety of such repositories. The disposal concepts investigated differ widely in the water content of the rock formations, the engineered barrier systems and the sorption capacities of the various materials. *To cite this article: W. Brewitz, U. Noseck, C. R. Physique 3 (2002) 879–889.*

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spent fuel disposal / rock salt / granitic rock / backfill / heating experiment / in-situ testing / performance assessment

Comportement à long terme de combustibles usés, placés dans des stockages

Résumé

La quantité de combustible consommé qui sera généré en Allemagne jusqu'en 2040 a été estimée à 8950 t_{hm}. À ce moment-là, 5500 t_{hm} de déchets vitrifiés de haut niveau auront été accumulés. Les deux types de déchets seront finalement enfouis dans un dépôt souterrain. Les roches-hôtes potentielles sont les roches salifères, le granit et les argilites. Jusqu'à maintenant, plusieurs études de validation de performances ont été réalisées pour identifier les principaux paramètres gouvernant la sûreté à long terme de tels dépôts. On a étudié des modèles de déposition très différents, selon la teneur en eau de la formation rocheuse, les systèmes de barrières développés et les capacités de sorption des divers matériaux. *Pour citer cet article: W. Brewitz, U. Noseck, C. R. Physique 3 (2002) 879–889.*

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* Correspondence and reprints.

E-mail address: brw@grs.de (W. Brewitz).

1. Introduction

Most recently the German Government came to an agreement with the German utilities on a limitation to the operation time of each nuclear power plant. This agreement forms the base of the new atomic act, which has passed the Federal Parliament in December 2001. In accordance the 19 German nuclear reactors are entitled to generate altogether another 2600 terawatt-hours, from 2000 onwards. This figure correlates with a mean full load over a lifetime of about 32 years. It is assumed that the last nuclear power plant will be shut down in the year 2022.

It was also agreed that after June 2005 no more spent fuel will be sent for reprocessing to France and Great Britain. This decision is based on a policy change in waste management away from reprocessing towards direct disposal of spent fuel. This is the background for the re-assessment of the radioactive waste volume in general and the quantities of HLW (high level waste) and SF (spent fuel) in particular, which have to be disposed of after the year 2030 at the latest (see Table 1). In the meantime there are two decisive steps to be made, the management of interim storage of SF and the site selection and construction of an underground repository.

By now, each electricity utility in Germany has applied for licensing of on-site interim storage for about 40 years. The technical concept of these facilities is based on the experience gained in dry storage of massive casks from the two central storage sites at Ahaus and Gorleben.

With respect to the final repository site under investigation at Gorleben the government imposed a moratorium of up to 10 years. In the meantime alternative sites will be identified and, as far as possible, investigated. In order to do this according to the advanced state of the art and also in a more public and transparent way the Federal Minister for the Environment, Nature Conservation and Reactor Safety has installed an independent expert group. Its main tasks are the definition of site selection criteria and the development of an iterative decision-making procedure incorporating public participation as far as possible. The group will make its recommendations by the end of 2002.

Despite of these management issues research and development work related to the final disposal of SF still proceeds under funding by the Federal Minister for Economics and Technology (BMW). Experiments in underground rock laboratories are focused on the development of disposal techniques and the determination of safety relevant parameters for construction, operation and licensing of heat generating radioactive waste.

2. Applied R&D for spent fuel disposal

The development of the disposal concept for spent fuel in Germany was initiated in the early eighties [2]. The subjects of this program were:

- development of disposal casks for fuel rods and structural parts including the necessary handling and conditioning techniques;
- in-situ handling and disposal test with inactive full-size Pollux casks;
- performance assessment for an underground repository in rock salt for all types of radioactive waste;
- laboratory research on material properties and performance, in particular under disposal conditions.

Table 1. Quantity of waste in Germany (actual stock + expected until 2030) [1]

Waste	Quantity		Total inventory [Bq]	Specific activity [Bq/t _{hm}]	Specific heat generation [W/t _{hm}]
	[m ³]	[t _{hm}]			
HAW	908	5550	$4.14 \cdot 10^{19}$	$7.45 \cdot 10^{15}$	1747
Spent Fuel	18 258	8947	$1.38 \cdot 10^{20}$	$1.54 \cdot 10^{16}$	1440
HTR	1890	10	$2.22 \cdot 10^{17}$	$2.2 \cdot 10^{16}$	3425

In order to maintain a high degree of flexibility two disposal concepts were investigated. The reference concept provides for the emplacement of Pollux casks in underground drifts. This cask meets all legal requirements for transport, interim storage, and final disposal. Its gross weight is about 65 t, the dimensions are 5.5 m length and 1.5 m in diameter and it can take up to 8 full-size LWR-fuel elements. The backup concept consists of Pollux canisters similar to those developed for HLW disposal in boreholes. Due to the smaller size and volume of such cans this concept requires more handling and cutting of the fuel rods.

The German Ministry of Education and Research (BMBF) signed responsible for the disposal simulation tests. Handling and loading of a 65 t Pollux cask was tested with a shaft hoisting mock up. It was demonstrated that the transport of loads up to 85 t to the underground is technically feasible and, what is very important in this respect, the cage taking canister and plateau carriage can be locked safely at the shaft installation for unloading. The underground handling and disposal tests were performed in the Asse salt mine. The exposure from scattered neutrons to be expected for the underground staff was modelled on the basis of experiments with neutron sources. The results indicates an increase of the dose by a factor of three. This has to be considered in the management of the disposal operation.

Most important was the mockup disposal test in backfilled underground drifts. The heating phase lasted from 1990 till 1999. It provided data for the characterization of the disposal near field such as temperature and stress distribution in backfill and rock mass, backfill compaction, rock mass deformation, gas generation and migration, and container material corrosion.

A number of performance assessment (PA) studies were focused on the disposal of all kinds of radioactive waste in a single repository. Different repository design and disposal strategies were incorporated into generic PA models. On the basis of conservative assumptions for all relevant safety parameters and processes the resulting individual doses were calculated for both the normal and the altered evolution scenarios. The effect of the most important parameters on the system performance was analyzed and the disposal concept for spent fuel was developed further and optimized.

This program paved the way for the decision by the Reactor Safety Commission (RSK) and subsequently for an amendment of the German Atomic Act making the direct disposal an alternative concept to HLW disposal. Today the R&D program of the Federal Minister of Economics and Technology is focused on the disposal of spent fuel not only in rock salt but also in other geological formations such as argillites and granites. In the framework of the Euratom Program GRS has participated in several international PA studies such as PAGIS, EVEREST and SPA [8]. Two host rock formations were considered: (i) rock salt representing basically dry conditions; and (ii) granite which stands, due to its fracture network, mostly for a wet environment.

2.1. System understanding by in-situ experiments

In order to investigate the heat-related effects in rock salt under realistic conditions a full-scaled simulation test was set up on the 800 m level in the Asse salt mine [3]. Two disposal galleries with three mockup Pollux casks each were separated by a 10 m wide rock pillar. The casks were fitted with electrical heaters of 6.4 kW each. The test site was monitored by pressure gauges, extensometers and various geophysical devices. The test was run over a period of eight years. Main objective was the validation of the thermo-hydraulic-mechanical (THM) models as well as the constitutive laws for the saliferous host rock and backfill. Porosity and permeability and their temporal changes are the most prominent parameters used for PA modelling.

Fig. 1 shows the temperature evolution at various locations of the test site. The maximum temperature at the canister surface was reached shortly after the start. Due to compaction of the backfill and subsequent increase of the heat conductivity the surface temperature decrease to a plateau of about 165 °C which is far below the critical limit of 220 °C for rock salt.

Over eight years a decrease in the backfill porosity of about 10%, from 35% to 25% was measured. Differences between experimental (blue curve) and calculated values, presented in Fig. 2, were analysed

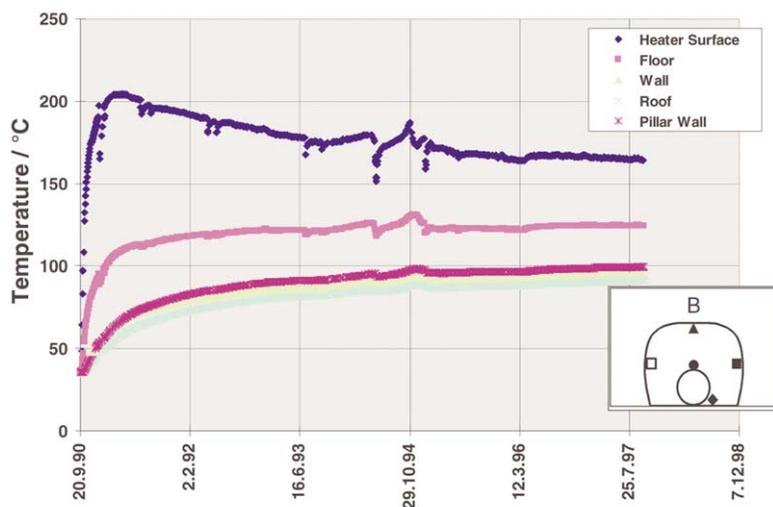


Figure 1. Temperature evolution at various locations of the thermal simulation test site.

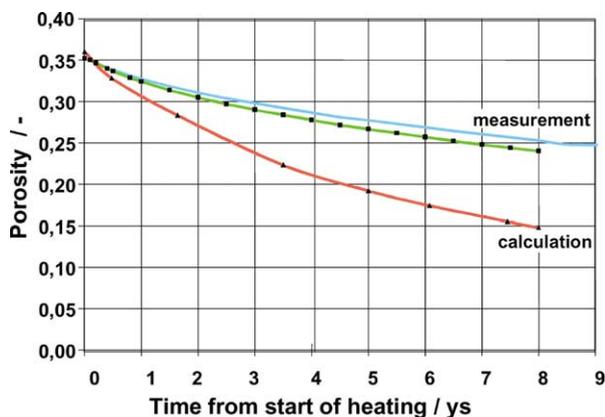


Figure 2. Porosity decrease versus time at the mockup test site.

in detail. It was shown that a simple 2D model overestimates the compaction which is quite decisive for any PA study. In a more realistic model, granular effects which contribute to some sort of self-support of the material, were considered. These effects hinder material compaction to some extent. The green curve describes, in agreement with the measured values, the real process fairly well, i.e. material compaction in the repository will take much longer than anticipated from laboratory experiments.

3. Performance assessment

In order to demonstrate the long-term safety of spent fuel disposal, predictive models were applied to normal case and altered evolution scenarios. For this purpose the computer programme EMOS (Fig. 3) was developed, which consists of separate modules describing the different parts of the repository system, i.e. the near field, the far field and the biosphere [4]. In each of these modules the safety-related processes of the repository system are included.

Main input data refer to the repository design and inventory, site-specific parameters characterising host rock and overburden, as well as to all relevant physical and chemical processes in the near and far field.

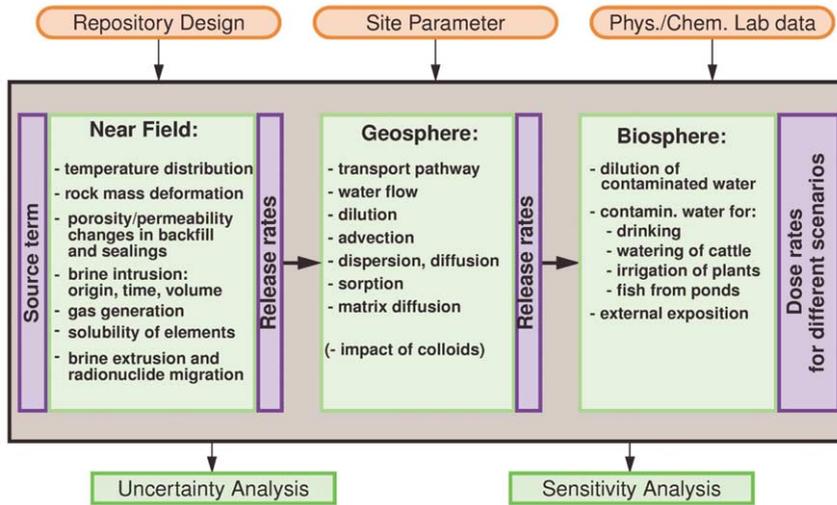


Figure 3. System of the integrated performance assessment code EMOS.

Table 2. Heat generating waste in different types of disposal containers

		Average container lifetime [y]	Heat generation [W] per container after					
			7 y	10 y	20 y	60 y	500 y	1000 y
HLW	steel can	5	2070	1657	1147	451	40	20
	large container	500	12 420	9943	6879	2706	240	120
Spent Fuel	small container	1000	3680	3020	2225	1100	205	115
	large container	500	9087	7460	5500	2720	510	290

3.1. Near-field modelling

Radionuclides are mobilised only if a sufficient volume of water is present and after the containers have corroded. Both processes, container failure and radionuclide release from the waste matrix, are included in the source term model. For thick-walled containers like the Pollux cask the failure can be described by an exponential law

$$n_C = 1 - \exp\left(-\frac{\tau}{\tau_{Cm}}\right) \quad (1)$$

with the number of failed containers n_C and the average container lifetime τ_{Cm} . Container life times estimated for different kind of containers are listed in Table 2.

The release rates for radionuclides are strongly dependent on the waste matrix and the geochemical environment of the near field. Concerning spent fuel three different fractions are dealt with in the source term model: the metal parts, the spent fuel matrix, and an instant release fraction of spent fuel. The degradation of each fraction is modelled with a constant rate:

- Some radionuclides are concentrated in the cladding and other metal parts of the fuel element (e.g. ^{14}C , ^{36}Cl , ^{60}Co). The release from all metal parts is assumed to last about 10^3 years based on corrosion rates of $10 \mu\text{m}/\text{year}$ for steel parts and $0.2 \mu\text{m}/\text{year}$ for the cladding. No credit is taken from a shielding effect of ZrO_2 layers.

- A significant part of radionuclides is accumulated in the gaps inside the rods and on the grain boundaries of the fuel. In particular I, Cl, Cs, Rb, and Zr are relatively quickly mobilized. About 5% of the inventory of these radionuclides are considered to be instantaneously released.
- Radionuclides of the spent fuel matrix will be released by a dissolution process which strongly depends on the redox potential. The modelling approach considers the dissolution of all nuclides together with UO_2 . Under reducing conditions the UO_2 matrix will be completely dissolved after 10^6 years, under oxidising conditions after 10^4 years. There is only a minor temperature dependence, which is not modelled.

The element specific mobilisation rates $R_{\text{LWR},x,i}$ for a radionuclide i are given by

$$R_{\text{LWR},x,i}(\tau) = n_C(\tau) \sum_x a_{x,i} r_{\text{LWR},x} \quad (2)$$

in which $a_{x,i}$ is the initial inventory of radionuclide i in fraction x (metal, spent fuel matrix, instantaneous release fraction) and $r_{\text{LWR},x}$ is the degradation rate for the material in fraction x . Additionally, some source-term models provide a γ - or α -dose-dependent dissolution rate, e.g. [5]. This considers the increase of oxidising substances by radiolysis, which leads to a greater dissolution rate of UO_2 .

For vitrified HLW it is assumed that radionuclide release from the matrix occurs congruently. The glass dissolution rate increases significantly with rising temperature. In some studies the release of a radionuclide i , $R_{\text{HLW},i}(\tau)$ is described as:

$$R_{\text{HLW},i}(\tau) = n_C(\tau) \frac{O_{\text{eff}} a_i}{m_0} r_{\text{HLW}} \exp\left(-\frac{Q_g}{R} \left(\frac{1}{T} - \frac{1}{T_R}\right)\right) \quad (3)$$

with an effective glass surface area O_{eff} , the inventory a_i of nuclide i , the initial mass of the glass matrix m_0 , the release rate r_{HLW} , the activation energy Q_g and the reference temperature T_R . Since new experimental results are now available, the source term is currently being adapted. The glass dissolution rate appears to be strongly dependent on the silica concentration of the solution becoming very slow at silica saturation.

3.1.1. Rock salt model

The German reference concept for a HLW/SF-repository in a salt dome is based on hundreds of disposal drifts or boreholes grouped together in different disposal sections. The specific concept for spent fuel is the emplacement of Pollux casks in disposal drifts and the backfilling with crushed salt. For near-field modelling the entire repository system is subdivided by a number of segment models.

For the normal case, as well as for the altered evolution scenario, rock salt properties and backfill properties are of utmost importance. In order to take most possible credit from the vast isolation potential of rock salt these material parameters have to be defined under normal and elevated temperature. The creep of rock salt strongly depends on the temperature, the minor constituents in the salt, the moisture and the stress field. The faster open voids are closed the earlier will the waste canisters become embedded in the formation. Once the self-healing process has ended the only scenario which may have to be considered is the so-called ‘human intrusion scenario’.

In rock salt the limiting parameter for the maximum heat load is the geochemical stability of some potash-bearing minerals which could release brines at temperatures above 220 °C. Fig. 4 shows the maximum temperatures to be reached in a HLW disposal borehole at the interface with rock salt and at the surface of a spent fuel cask in the centre of a backfilled disposal gallery. The temperature rise in the overall repository is considerably lower. For the calculation an interim storage time of 40 years for HLW and 30 years for spent fuel and repository closure after 50 years were assumed. Longer interim storage and less loading of the disposal casks will lead to a lower peak temperature for both HLW and spent fuel.

If no site-specific data are available the possibility of undetected brine pockets has to be considered in any generic radionuclide release model for rock salt. Such pockets can lead to limited brine inflow into the

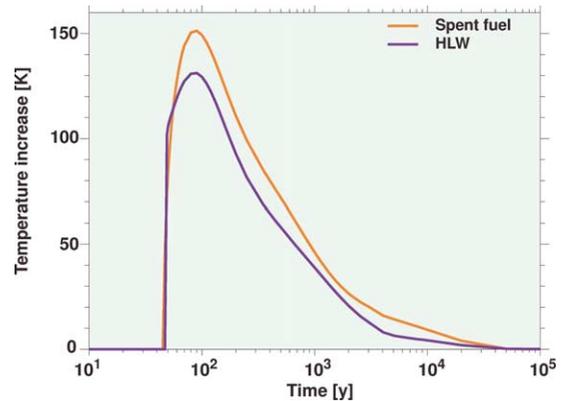


Figure 4. Peak temperatures reached in a HLW/SF-repository in rock salt.

repository. Under extremely unfavourable conditions anhydrite layers connecting the repository with the top of the salt dome may become a pathway for an unlimited brine intrusion. In such a scenario brine will enter the backfilled sections some time after closure of the repository. After all open voids are filled with brine radionuclides are mobilised and transported by advective or convective flow and diffusion. The advective transport is caused by the convergence of the cavities, convective transport is mainly due to temperature gradients. Temperature effects and solubility limits are taken into account in each section of the repository. The contaminated brine is extruded from the salt dome into the covering rock strata, quarternary and tertiary sediments. For total system performance modelling radionuclide release rates from the near field are input for the far-field modules.

3.1.2. Granite model

Due to the presence of water in granitic rocks nearly all concepts for spent fuel disposal are based on compacted bentonite as the geotechnical barrier around the containers. It reduces water migration, stabilizes the geochemical conditions in the near field for long periods of time, and has a high sorption capacity for most radionuclides. After radionuclide mobilisation, precipitation can occur, which is described by solubility limits. Due to the low permeability of water-saturated bentonite radionuclide transport through the bentonite buffer is diffusion-controlled. In granite the excavation disturbed zone is a potential pathway for radionuclide transport. In direct contact with the bentonite buffer, it is considered as an interface between the near field and the fracture network in the rock mass. The resaturation of the bentonite after repository closure and the effects of gas pressure build-up caused by metal corrosion are important processes, which are under investigation on national and international level.

3.2. Results from selected PA studies

For performance assessment studies, advanced conceptual and numerical models were used and the latest data were collected for specific disposal concepts. The results reveal important aspects for future experimental and analytical research.

The deterministic modelling of a brine intrusion scenario in rock salt is based on an amount of 25 000 t spent fuel disposed of in Pollux casks [6]. The earliest individual dose rates greater than 10^{-9} Sv/y occur 5 500 years after repository closure. This is due to the groundwater travel time in the covering rock strata of more than 1 000 years. The maximum dose rate of 10^{-5} Sv/y after 10 500 years correlates mainly with the release of ^{129}I and its low retardation and long half-life. Other important radionuclides in the first 10^5 years are ^{79}Se and ^{135}Cs . Most actinides are more strongly retarded and therefore reduced by dispersion and radioactive decay. However, in the time frame beyond 10^5 years ^{237}Np contributes significantly to the total dose rate and therefore is a good indicator for the long-term performance of the total system (Fig. 5).

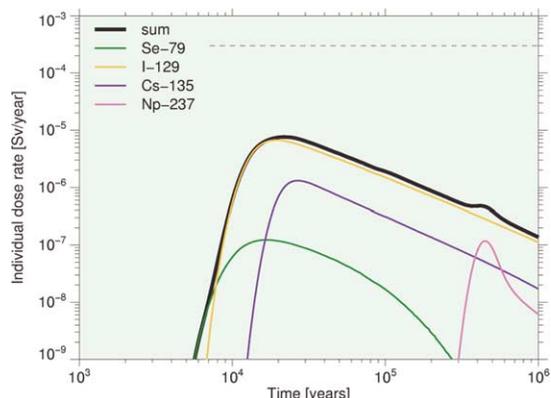


Figure 5. Total dose rates calculated for a brine intrusion scenario in a SF-repository in rock salt.

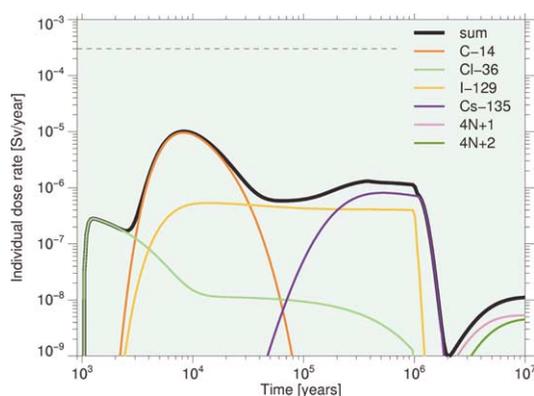


Figure 6. Total dose rates calculated for the reference case of a German SF-repository in granite.

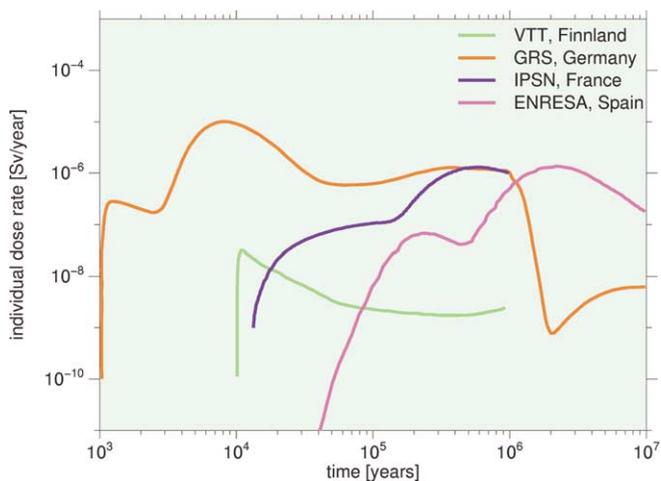


Figure 7. Total dose rates calculated for SF-repositories in granite (EU-project SPA).

The reference case for a SF-repository in crystalline formations [7] is also based on a spent fuel inventory of 25 000 t_{hm} packed in thick-walled stainless steel containers. The calculated dose rate also results from weakly- or non-sorbing radionuclides ^{14}C , ^{36}Cl , ^{129}I and ^{79}Se (Fig. 6). The near field and the rock mass are efficient barriers for the actinides and their daughter products e.g. ^{242}Pu , ^{234}U , ^{230}Th , ^{226}Ra , and ^{237}Np . The arrival time of the first radionuclides in the biosphere is governed by the container lifetime of 1000 years. For non-sorbing radionuclides the assumed geosphere conditions represent nearly no barrier. For the groundwater travel times in the far field of the repository only 50 years were taken into account.

The different PA studies on SF-repositories in granite were evaluated and compared in the EU-project “Spent Fuel Performance Assessment” (SPA) [8]. The calculated dose rates for the reference cases of the four participating institutions are illustrated in Fig. 7. Differences mainly reflect the different assumptions for container life time and for radionuclide retention and dilution in the far field.

In the Finnish analysis only one container is assumed to fail after 10 000 years, whereas in all other studies containers fail after about 1000 years. This becomes obvious in the initial radionuclide release into the biosphere in the studies of VTT and GRS. Both are based on fast radionuclide transport in the far field. In the other two studies much longer groundwater travel times are considered.

Transuranic elements play a minor role in the studies of ENRESA and GRS, whereas they are dominating after 10^5 years in the IPSN study. Sorption processes and the respective modelling approaches account for these differences. In comparison the double porosity model with matrix diffusion and linear sorption results in much longer travel times for strong sorbing transuranic elements than the equivalent porous medium model with similar K_d -values. Additionally, higher solubility limits in particular for U, Th and Np contribute to the significantly higher dose rate of the transuranic elements in the IPSN study.

4. Conclusions and outlook

The analytical tools and methodologies have been developed to a high standard giving a good indication on the long-term performance of the total repository system. Almost site independent are the effects and processes in the near field which contribute substantially to waste isolation and nuclide retention. As demonstrated, more precise and validated data about the properties and behaviour of the technical and geotechnical barriers may help to improve the long-term safety of the disposal concepts. Some processes related to chemical interactions between waste, waste containers and the intruding groundwater have to be understood more thoroughly in order to reduce over conservative assumptions.

With respect to the far field groundwater travel time and sorption capacities of the different rock formations are of utmost importance. These parameters can only be derived from site-specific investigations. From this point of view the modelling of generic disposal concepts is subject to a certain degree of uncertainty. In particular, those disposal formations in which groundwater is present require a detailed survey of all geological features which may contribute to greater radionuclide retention. In this context basic research needs are:

- better understanding of gas generation processes and the impact on disposal system stability and radionuclide transport;
- detailed characterisation and gradual optimization of near-field barriers;
- nuclide solubility and behaviour in specific geochemical environments, coupling of geochemistry and transport;
- thermal-hydraulic-mechanic behaviour of the repository near field in granite and clay formations;
- harmonisation of submodels and data;
- incorporation of extended features events and processes (FEPs) and flow entries into the different PA models;
- more precise data on nuclide migration in clay and other rock matrices;
- extended data base for long-term convergence of underground voids and backfill compaction in rock salt;
- more data for detailed hydrogeological modelling of hard rock formations including discretization of main faults in granite;
- more systematic treatment and reduction of data uncertainties.

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Discussion

Question from C. Fairhurst

How is human intrusion considered in Performance Assessment for repositories in Germany?

Reply from W. Brewitz and U. Noseck

So far 'human intrusion' was and is not a great issue in Germany. With respect to the site investigation of the Gorleben salt dome a total system performance analysis has not been initiated yet. That is why this subject was considered in basic research only. In non-site-specific and generic PA solutions mining was identified as a realistic intrusion scenario for rock salt formations. In a very first model for HLW and ILW disposal the consequences have been calculated resulting from the exploration and construction of an underground gas storage and of solution salt mining. The modelling results have proved that such a scenario contributes to the dose rates in the order of magnitude calculated for any other brine intrusion scenario.

Question from C. Madic

In case of the disposal of spent waste in rock salt, during the 'heat period', the salt with brine inclusions will move towards the heat source. So, water will come in contact with the canister. Did you take into account this phenomenon in your modelisation of the disposal of spent fuel in this rock salt?

Reply from W. Brewitz and U. Noseck

In the Asse salt mine a number of heating tests have been performed in the past 20 years. An outstanding experiment was the so called 'brine migration test' performed by GRS and Battelle/ONWI in 1984/85. The set up provided full heat load to the rock salt. Minor traces of brines were detected in the order of 70 g per meter of heated borehole. Small volumes of hydrogen were also measured indicating the corrosion of some parts of the steel liners. These findings were supported by a numerical model on canister corrosion and gas generation based on laboratory and in situ data.

Question from J. Dercourt

Concerning 'long-term performance', the canisters containing nuclear material emit energy and raise the temperature; what would be the consequences for the ductility of the salt in your experiments?

Reply from W. Brewitz and U. Noseck

The creep behaviour is the predominant parameter governing the convergence and subsequently the closure of open voids in underground repositories in rock salt formations. Under heat load the creep is

strongly accelerated which leads very soon to an almost complete enclosure of high level waste canisters in boreholes. With respect to seals and backfill in boreholes and disposal galleries this process is the driving force for faster material compaction. In general it is a fact that the rheological properties of rock salt are most favourable for the disposal of heat generating radioactive waste.

Question from R. Dautray

In which German mines (Morsleben, Asse, Konrad) have you made your experiments? Unfortunately, we are unable to use suitable French mines to make similar tests.

Reply from W. Brewitz and U. Noseck

The disused salt mine Asse was selected as an underground laboratory and a demonstration facility for low and intermediate level waste disposal at a very early stage of the German radioactive waste management program. Technical and rock mechanical experiences gained in more than 30 years of operation provided the basis for any further considerations of radioactive waste disposal not in rock salt but also in any other rock formation. In the mid-seventies it became clear that the Asse had not the potential for becoming a licensed underground repository. In addition it became obvious that a number of parameters can only be investigated at the specific site pre-selected as repository. That is why research and development was reduced to a minimum when the exploration work started at Gorleben. Today the Asse is being backfilled setting an example for any underground repository being shut down in the future.

Remark from B. Tissot

You have insisted, rightly, on the importance of the parameters which depend on the geological site. Many people think that all clays are equivalent, or all granites the same. This could be a source of considerable error, and I am glad to hear your views.

Reply from W. Brewitz and U. Noseck

I can only confirm this. Despite a good understanding of the basic rock mass properties and the geoprocesses governing the post-operational safety it is a must to perform an in-depth site characterisation program. Quite a number of parameters are site specific or depend to some extent on the design of the repository with its various components such as shafts, galleries, rock pillars and its future backfilling and seals. How these fit together and how the system works can only get checked at the site itself. The suitability of a site can never be proved by desk studies and off-site experiments alone.

Remark from P. Toulhoat

Concerning your source term, we have some recent evidence that the cladding will not be able to resist the pressure induced by alpha decay. After some thousand years, the rod can be turned into powder.

Reply from W. Brewitz and U. Noseck

The German PA studies for SF disposal are based on simple and conservative assumptions. Since uptake of hydrogen and stress induced cracking will lead to a successive disintegration of the claddings it was assumed that after the failure of the disposal containers, complete corrosion of SF will take place, including the corrosion of the fuel pellets. In case of salt brines this makes 250 years for the release of all radionuclides from the zircaloy claddings.